

**WE CLAIM:**

1. A carbon-based hydrogen storage composition comprising a pillared carbon material doped with a metal.
2. The composition of claim 1, wherein the metal is selected from alkali metals, alkaline-earth metals, and combinations thereof.
3. The composition of claim 2, wherein the metal is selected from Li, Na, K, Be, Mg, Ca and combinations thereof.
4. The composition of claim 1, wherein the carbon material is selected from graphite, graphene, carbon nanostructures, and combinations thereof.
5. The composition of claim 4, wherein the carbon material is selected from graphite, graphene, carbon nanofibers, carbon nanocells, carbon nanobarrels, multi-wall carbon nanotubes, single-wall carbon nanotubes and combinations thereof.
6. The composition of claim 1, further comprising an impurity or an additive.
7. The composition of claim 6, wherein the impurity or additive is selected from B, N and combinations thereof.

8. The composition of claim 1, wherein the atomic proportion of the metal to the carbon atoms in the pillared carbon material is in the range of about 1:3 to about 1:24.
9. The composition of claim 8, wherein the atomic proportion of the metal to the carbon atoms in the pillared carbon material is in the range of about 1:3 to about 1:8.
10. The composition of claim 9, wherein the atomic proportion of the metal to the carbon atoms in the pillared carbon material is in the range of about 1:3 to about 1:6.
11. The composition of claim 1, wherein the pillared carbon material has an interlayer or intertube distance in the range of about 6 Å to about 12 Å.
12. The composition of claim 10, wherein the pillared carbon material has an interlayer or intertube distance in the range of about 6 Å to about 12 Å.
13. The composition of claim 12, having a molecular hydrogen storage capacity of at least about 3 wt.% at 25°C and a pressure of about 10 bar.
14. The composition of claim 13, having a molecular hydrogen storage capacity of at least about 6.5 wt.% at 25°C and a pressure of about 10 bar.
15. A hydrogen storage system comprising a carbon-based composition according to claim 1.

16. In a hydrogen storage system, wherein a carbon material is used to store hydrogen, the improvement which comprises employing a carbon-based composition comprising a pillared carbon material doped with a metal.
17. A method of making a carbon-based hydrogen storage composition, comprising
- providing a solvated alkali metal containing organic ligands;
  - combining a carbon material with the solvated alkali metal containing organic ligands to form a carbon material co-intercalated with alkali metal cations containing organic ligands;
  - carrying out a reaction between the organic ligands and the carbon material to form a pillared carbon material; and
  - doping the pillared carbon material with a metal.
18. The method of claim 17, wherein the alkali metal of the solvated alkali metal cation is selected from Li, Na, K, and combinations thereof.
19. The method of claim 17, wherein the doped metal is selected from alkali metals, alkaline-earth metals, and combinations thereof.
20. The method of claim 19, wherein the doped metal is selected from Li, Na, K, Be, Mg, Ca and combinations thereof.
21. The method of claim 17, wherein the carbon material is selected from graphite, graphene, carbon nanostructures, and combinations thereof.

22. The method of claim 21, wherein the carbon material is selected from graphite, graphene, carbon nanofibers, carbon nanocells, carbon nanobarrels, multi-wall carbon nanotubes, single-wall carbon nanotubes and combinations thereof.

23. The method of claim 17, wherein said organic ligand solvated alkali metal cation comprises an organic solvent selected from heterocyclic solvents.

24. The method according to claim 23, wherein said organic solvent is a cyclic ether compound.

25. The method according to claim 24, wherein said organic solvent is 2,5-dihydrofuran.

26. The method according to claim 17, wherein said doping includes intercalation of the metal and ball milling of the pillared carbon material.

27. The method of claim 17, wherein the carbon material further comprises an impurity or an additive.

28. The method of claim 27, wherein the impurity or additive is selected from B, N and combinations thereof.

29. The method of claim 17, wherein the atomic proportion of the metal to the carbon atoms in the pillared carbon material is in the range of about 1:3 to about 1:24.

30. The method of claim 29, wherein the atomic proportion of the metal to the carbon atoms in the pillared carbon material is in the range of about 1:3 to about 1:8.

31. The method of claim 30, wherein the atomic proportion of the metal to the carbon atoms in the pillared carbon material is in the range of about 1:3 to about 1:6.
32. The method of claim 17, wherein the pillared carbon material has an interlayer or intertube distance in the range of about 6 Å to about 12 Å.
33. The method of claim 31, wherein the pillared carbon material has an interlayer or intertube distance in the range of about 6 Å to about 12 Å.
34. The method of claim 33, wherein the carbon-based composition has a molecular hydrogen storage capacity of at least about 3 wt.% at 25°C and a pressure of about 10 bar.
35. The method of claim 34, wherein the carbon-based composition has a molecular hydrogen storage capacity of at least about 6.5 wt.% at 25°C and a pressure of about 10 bar.
36. In a method of making a hydrogen storage device, wherein a carbon material is used to store hydrogen, the improvement which comprises employing a carbon-based composition comprising a pillared carbon material doped with a metal.